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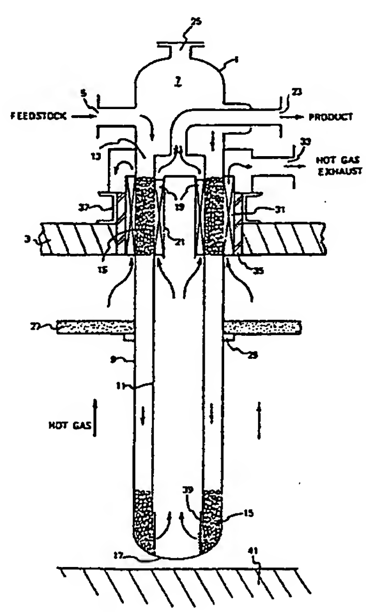
(54) **Catalytic reactor**

(57) A catalytic reactor of a coaxial double-tube construction is provided with a fluid reactant inlet (5) and a fluid reactant outlet (2) at one end thereof. The other end of outer tube (9) is closed (at 17), and projects into a heating vessel, a fluid reactant fed through inlet (5) flows through an annular space formed between the tubes (9) and (11) which is packed with a catalyst towards said other end, changes direction, and then flows through the inner tube (11) towards the outlet (23). A heating gas flows along the outer surface of the outer tube (9) from said other end towards said one end. Heat recovery fins (19) are provided within the inner tube (11) so as to project from the inner surface of the inner tube.

In addition further heat recovery fins (31) project from an outer face of the outer tube to collect heat from gas exhausted from the furnace.

Gas-permeable ceramic wall (27) may be provided to enhance heating by radiation.



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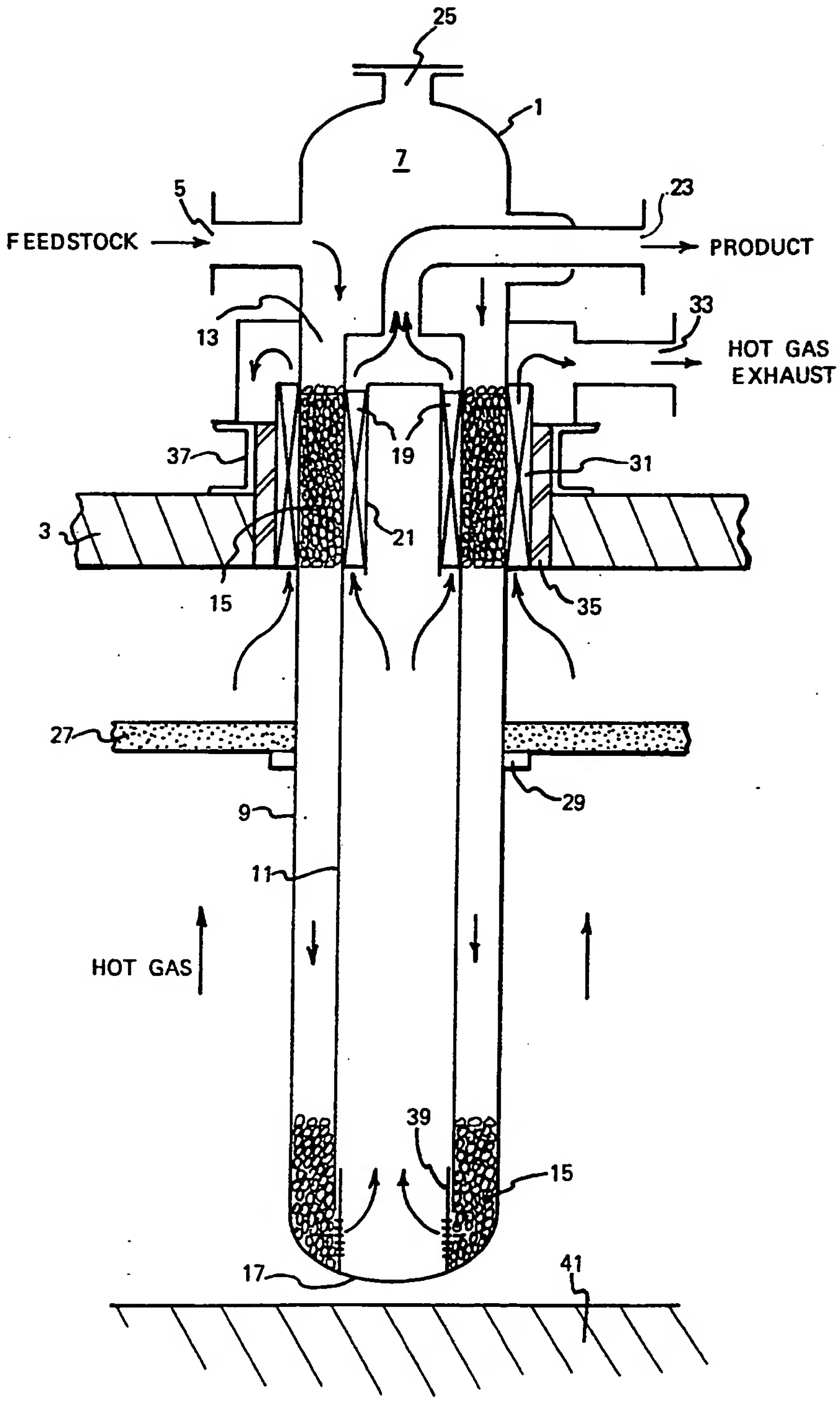
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DESCRIPTION

CATALYTIC REACTOR

The present invention relates to a catalytic reactor. More specifically, the present invention relates to an apparatus for an endothermic reaction, such as a steam reforming reaction for reforming hydrocarbons, capable of operating at a high heat recovery efficiency and/or a low energy consumption.

There has been proposed a steam reforming reactor for a hydrogen generator, internally provided with a catalyst for high-temperature reaction using heat supplied from an external heat source. This steam reforming reactor employs a double-tube reaction structure having an outer tube and an inner tube disposed coaxially with the outer tube to achieve efficient heat exchange, to miniaturize the arrangement, and to form an annular space packed with a catalyst between the outer and inner tubes. A high-temperature reaction product gas flows through the inner tube. The employment of the double-tube reaction structure enables transfer of the sensible heat of the reaction product gas flowing through the inner tube, after passing the catalyst layer heated at a high temperature, through the wall of the inner tube to the catalyst layer, reduction of the capacity of a heat recovery equipment comprising heat exchangers and

the like provided on the downstream of the steam reforming reactor, and reduction of heat dissipation because the reaction product gas is discharged from the outlet of the steam reforming reactor at a comparatively low temperature.

Japanese Patent Provisional Publication (Kokai) No. 59-16536 proposes a double-tube reactor capable of operating at a higher heat utilization. This double-tube reactor has a double-tube structure having an inner tube internally provided with a radiator having heat radiative surfaces. This radiator is for the purpose of heat transfer to the wall of the inner tube by radiation as well as by convection from the sensible heat of the reaction product gas, because the radiator is heated by convection. The reaction product gas flowed out of the catalyst layer goes upward through the inner tube internally provided with the radiator.

There are instances where sufficient heat exchange cannot be expected from the foregoing reactor incorporating the radiator.

As regards heating the reactor, heat is transferred mainly by radiation from a high-temperature combustion gas to the reactor in a heating furnace, i.e., a typical heating vessel, in which the reactor is installed. The exhaust gas,

namely, the combustion gas which has transferred its heat to the reactor allowing its temperature to drop to a level not high enough to transfer heat by radiation, is passed through an exhaust gas passage to recover the sensible heat of the exhaust gas by, for example, a heat exchanger comprising folded pipes provided within the exhaust gas passage. In such a case, the reactant (feedstock) gas may be passed for preheating through the heat exchanger to feed the reactant gas to the reactor, which, however, requires unavoidably complicated piping. Discharge of the exhaust gas still having a high temperature from the reactor increases heat dissipation, which reduces the overall efficiency of the reactor significantly.

It is an object of the present invention to provide means capable of avoiding the foregoing problems, having a more simplified construction capable of improving the efficiency of heat exchange, and capable of reducing pressure loss in the reactant fluid and/or the combustion gas to the least possible extent to reduce power necessary for boosting-up feed.

The present invention employs heat recovery means, such as fins or projections, bonded integrally to the relevant heat transfer wall by metallurgy so as to project from the heat transfer wall to enhance heat transfer convection for the effective recovery of the

sensible heat of the reaction product gas and/or the exhaust gas.

The present invention comprises a catalytic reactor of a double-tube construction provided at one end thereof with a fluid reactant inlet and a fluid reactant outlet, wherein the other end of an outer tube is closed, a portion of a double-tube structure other than a portion of said one end projects into a heating vessel, whereby a fluid reactant fed through the fluid reactant inlet flows through an annular space formed between the outer tube and an inner tube and packed with a catalyst toward said other end, changes the flowing direction at said other end, and then flows through the interior of the inner tube towards the outlet, and whereby a heating gas flows along the outer surface of the outer tube from said other end towards said one end, and heat recovery means is provided within the inner tube so as to project from the inner surface of the inner tube.

The invention is further described, by way of example, with reference to the accompanying drawing which is a schematic longitudinal sectional view of a catalytic reactor in a preferred embodiment according to the present invention.

The catalytic reactor shown in the drawing is disposed within a heating furnace.

The catalytic reactor 1 is secured on the upper wall (ceiling) 3 of the furnace. In this embodiment, a mixed material (feedstock) gas, namely, a mixture of hydrocarbons capable of steam reforming reaction and steam, is fed through a catalyst layer and is heated by the heat supplied from the heating furnace to convert the gas mixture mainly into hydrogen and carbon dioxide.

The mixed gas is fed through an inlet 5 and the internal space 7 of a top cap to an annular space 13 formed between an outer tube (cylinder) 9 and an inner tube (cylinder) 11 and packed with a reforming catalyst 15 (in the drawing the reforming catalyst in the middle portion of the annular space is omitted). The upper portion of the catalyst layer serves mainly for preheating the gas mixture and the lower portion other than the upper portion of the same serves for reaction.

The gas mixture is reformed while flowing through the catalyst layer and is converted into a reaction product gas before arriving at the bottom 17 of the reactor tube. Then, the reaction product gas flows upwardly through the interior of the inner tube. Plate fins 19 are bonded integrally in parallel to the flow passage to the inner surface of a portion of the inner tube corresponding to the portion of the

catalyst layer serving for preheating, by metallurgical means. Welding or brazing is suitable metallurgical means for integrally bonding metallic plate fins having a high thermal conductivity to the inner tube. Metallurgically integral bonding is employed because such bonding is superior to simple contact connection in heat transferring performance. A method of integrally bonding fins formed by corrugating thin plates, such as thin stainless steel plates, to the inner surface of the inner tube by welding or brazing, and providing a dummy tube 21, which may be formed of the same material as the fins, may be employed to increase the heating surface area of the fins maintaining the pressure loss at the lowest possible degree. In this case, vacuum brazing is suitable because the fins are formed of thin plates. An object of the present invention being the enhancement of heat exchange efficiency without increasing pressure loss, such thin corrugated plate fins are suitable.

One end of the dummy tube 21, the upper end, preferably, is closed so that the reaction product gas may flow only through passages between the plate fins.

The sensible heat of the reaction product gas is transferred through the plate fins and the wall of the inner tube to the portion of the catalyst layer

serving for preheating to heat the gas mixture flowing downward through the catalyst layer by the sensible heat, so that the temperature of the reaction product gas drops. After thus decreasing in temperature, the reaction product gas is discharged from the outlet 23 of the reactor.

Then, ordinarily, the reaction product gas discharged from the catalytic reactor is processed by a carbon monoxide converter and a pressure swing adsorber to increase the hydrogen purity of the reaction product gas for desired purposes, such as the feed to hydrogen electrodes of fuel cells. In Fig.1, indicated at 25 is an opening for supplying the catalyst to pack the reactor therewith.

The catalytic reactor shown in Fig.1 is provided with a gas-permeable wall 27 of a ceramic material to utilize the radiant heat of the combustion gas effectively. The ceramic plate wall 27 is supported by the reactor tube at an approximately central position with respect to its length so as to surround the reactor tube and so as to partition the internal space of the furnace into upper and lower sections. Indicated at 29 is a support for supporting the ceramic plate.

In the lower section below the wall 27, i.e. radiation heat transfer spaces, heat of the combustion

gas is transferred from the combustion gas to the reactor tube mainly by radiation. Accordingly, it is inadvisable to provide plate fins in the lower section. After transferring heat by radiation, the combustion gas passes through the gas-permeable ceramic wall heating the ceramic wall by its sensible heat. Then, the ceramic wall radiates heat into the lower section to heat the portion of the reactor tube projecting downward from the ceramic wall.

The gas-permeable ceramic wall may be substituted by a gas-permeable wall formed of any material other than a ceramic, such as a material at least partially formed of a metallic material, provided that the material is heat-resistant and gas-permeable, and capable of effectively returning heat into the radiation heat transfer space and allowing the combustion gas to pass through the gas-permeable wall at a low pressure loss.

Although the temperature of the combustion gas passed through the gas-permeable wall is not high enough for heat transfer by radiation, the combustion gas still has a sufficient sensible heat at this stage. To use this heat of the combustion gas effectively for preheating the gas mixture, heat recovery means for recovering heat by convection, namely, plate fins 31 similar to the aforementioned

plate fins 19, is bonded integrally to the outer suface of the reaction tube by metallurgical means. Since a tube surrounding the plate fins 31 need not contribute to heat transfer, it may be provided by simple wrapping.

The section of the heating vessel above the gas-permeable material is designated as a convection heat transfer space as contrasted with a radiation heat transfer space.

The ratio of vertical thickness between the two heat transfer spaces is varied mainly according to the temperature of the heating gas.

A gas heated by a heat source, which may be one utilizing combustion outside the heating vessel, as well as a gas of a combustion furnace formed in the radiation heat transfer space of the heating vessel, may be supplied into the radiation heat transfer space followed by flowing through the gas-permeable wall into the convection heat transfer space.

The reactor further comprises an exhaust gas outlet 33, a heat insulator 35, a support 37 formed by steel sections, a partition tube 39 provided within the double-tube structure, and the bottom wall (floor) 41 of the furnace.

The partition tube 39 prevents the catalyst packed in the annular space from entering the inner tube 11.

In this embodiment, the partition tube 39 is fixed coaxially with the inner tube 11 to the bottom 17 of the outer tube 9 so as to be axially movable relative to the inner tube along the inner or outer surface of the inner tube. A portion of the partition tube 39 between the lower end of the inner tube 11 and the bottom 17 of the outer tube 9 is perforated to enable the reaction product gas to flow from the annular space through the perforated portion into the inner tube. Since the partition tube 39 is fixed to the outer tube, the movement of the inner and outer tubes relative to each other attributable to the difference of thermal expansion between them can readily be absorbed.

The sensible heat of the reaction product gas and that of the combustion gas can effectively be recovered by both sets of plate fins 21,31 provided on the inner surface of the inner tube 11 of the reaction tube and on the outer surface of the outer tube 9 of the reaction tube. According to the illustrated embodiment of the present invention, the plate fins are provided both on the inner surface of the inner tube of the reaction tube and on the outer surface of the outer tube of the reaction tube as stated above; plate fins may, however, be provided, if necessary, on the inner and outer surfaces of the inner tube of the

reaction tube and/or on a portion or whole area of the inner and outer surfaces of the outer tube of the reaction tube taking into consideration calculated thermodynamic advantages and structural feasiblity.

Effect of the Invention

As is apparent from the foregoing description, the catalytic reactor according to the present invention provided in the heating vessel is provided with heat recovery means, in particular heat transfer means, such as heat exchangers provided with corrugated plate fins which are effective on increasing the heat transferring area, are provided at the necessary portions of the inner tube and/or the outer tube of the reactor tube thereof. Thus, the sensible heat of the reaction product gas and that of the combustion gas can sufficiently be used for preheating the gas mixture (feedstock). Consequently, the heat recovery efficiency is increased, the overall heat exchange efficiency of the heating surface is improved and the reactor can be formed in a compact construction. Furthermore, since the reactant fluid and/or the combustion gas is able to flow at a comparatively small pressure loss, the power necessary for the forced flow of the reactant fluid and/or the combustion gas can be reduced.

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CLAIMS

1. A catalytic reactor of a coaxial double-tube construction provided at one end thereof with a fluid reactant inlet and a fluid reactant outlet, wherein the other end of an outer tube is closed, a portion of a double-tube structure other than a portion of said one end projects into a heating vessel, whereby a fluid reactant fed through the fluid reactant inlet flows through an annular space formed between the outer tube and an inner tube and packed with a catalyst towards said other end, changes the direction of flow at the other end, and then flows through the interior of the inner tube towards the outlet, and whereby a heating gas flows along the outer surface of the outer tube from said other end towards said one end, and heat recovery means are provided within the inner tube so as to project from the inner surface of the inner tube.

2. A catalytic reactor according to claim 1, wherein a heating gas outlet through which the heating gas flows out of the heating vessel is formed by an annular space coaxial with the double-tube structure, provided between the outer wall of a portion of the outer tube near said one end of the double-tube structure and the wall of the heating vessel on the side of said one end of the double-tube structure,

through which heating vessel wall the double-tube structure projects.

3. A catalytic reactor according to claim 1 or 2, wherein heat recovery means projects from the outer surface of the outer tube.

4. A catalytic reactor according to claim 1, 2 or 3, wherein the heat recovery means is provided at a position corresponding to the heating gas outlet.

5. A catalytic reactor according to any of claims 1 to 4, wherein the heat recovery means comprises plate fins forming flow paths parallel to the axis of the tubes.

6. A catalytic reactor according to any of claims 1 to 5, wherein the internal space of the heating vessel is divided into a radiative heat transfer space on the side towards said other end of the double-tube structure and a convection heat transfer space on the side towards said one end of the double-tube structure by a gas-permeable wall through which said other end of the double-tube structure extends away from the wall on the side of said one end of the heating vessel.

7. A catalytic reactor constructed, arranged and adapted to operate substantially as herein described with reference to and as illustrated in the accompanying drawing.

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